Phase-ordering dynamics for the onset of a center manifold

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We derive the correlation of spatially coupled subsystems in the case in which a dissipative organization is sustained in an open reactive system which operates far from equilibrium. This phenomenon is viewed in the context of phase-ordering dynamics of thermodynamically unstable systems, where the organization is achieved by amplification of nonequilibrium fluctuations (as in spinodal decompositions and in nucleation processes). Thus we show how the correlation of subsystems leads to the contraction of phase space to a center manifold and to the propagation of fluctuations along the center manifold in order to reach the dissipative structure.

I. INTRODUCTION

The onset of far-from-equilibrium organizations in dissipative systems is a problem of active current interest, especially since such dissipative structures do not enjoy the status of permanence and stability that equilibrium structures have.[1-4] The cooperativity among spatially coupled subsystems producing collective fluctuations on long-lifetime modes is poorly understood in the context of dissipative structures.[4] Relevant areas of research are, for example, the nature of intrinsic noise sources in the order-parameter evolution for the onset of convective patterns,[5,6] intrinsic fluctuations as triggers of dissipative structures in open reactive systems,[4] correlation of subsystems in spinodal decompositions,[7,8] etc.

The question of permanence and stability of far-from-equilibrium organizations has been answered in some specific instances, for example, when the organization lies on a locally attractive and locally invariant portion of phase space known as the center manifold (CM).[3,4] Essentially, this surface is obtained by a mean-field approach with a free-energy functional including third-order terms with mode coupling between order parameters and fast-relaxing degrees of freedom. The order parameters which arise beyond the critical instability are the CM coordinates. The surface represents the statistical enslavement of fast-relaxing modes to the order parameters.

The nature of the cooperativity involving a large number of particles in the system and leading to the CM contraction of phase space is far from being understood. The related question which shall be addressed in this paper is how does the amplification and propagation of fluctuations along the CM occur in order to reach the attractor. The transient for this process is experimentally accessible, at least in some instances,[5,6] thus the predictive value of our theory can be tested. We shall demonstrate that the onset is not due to a highly improbable large fluctuation[1,2] but rather to the propagation and amplification of a "primeval" fluctuation along the CM, where the attractor lies. The origin of this initial fluctuation will be given later. The process involves cooperativity of spatially coupled subsystems as defined by the following two properties.

(i) The onset of a CM determines a decomposition of the system of volume V into cells of virtual volume W, so that collective fluctuations scale with the small parameter $W/V=L^{-1}$.

(ii) Subsystems whose macrostates lie in the CM are regarded as "organized," and organization is induced from one subsystem onto another. Moreover, each "information carrier" which lies in the CM, excluding the attractor, has a finite lifetime which depends on the distribution of probability about the CM.

Thus the process leading to evolution of fluctuations towards the attractor can be studied by means of a model consisting of organization carriers which can reproduce, mutate into one another, and decay subject to the constraint of constant overall population. The details of this model and its implementation and computational effectivity in order to yield transients or induction periods for the onset of nonequilibrium organizations will be derived in Secs. II and III.

The analysis of the propagation and amplification of fluctuations along the CM beyond a dynamic instability is in the spirit of studies of the decay of metastable states due to nucleation effects in alloys (spinodal decomposition) and ferromagnets. The interaction of spatially correlated subsystems leading to the conservation of the order parameter has already been considered in the context of spinodal decompositions and other nucleation processes. Our analysis is analogous in so far as we consider the decay of a metastable state by amplification of nonequilibrium fluctuations among spatially coupled cells.

II. CENTER MANIFOLD REDUCTION

As stated in Sec. I, a CM is a surface in the space of collective modes of many-body systems. This surface accounts for the phase-space contraction given by the adiabatic following of certain subordinated modes to the order parameters in the critical regime. A clear illustration of this is the transition to a convective pattern in a Rayleigh-Bénard cell, the CM is bilinear in the slowly varying Fourier modes, and the CM coefficients are obtained by taking the projection of the nonlinear terms in the Boussinesq flow onto the subordinated Fourier modes (see, for example, Ref. 5).

Thus the collective fluctuations introduce a random-source term on the order-parameter equation. The effective virtual volume to which such fluctuations are confined is determined by the dynamics of the particular unfolding. This is done as follows. First, an integration of the generalized master equation for the probability density functional $P=P(\mathbf{X},t)$ ($\mathbf{X}$ is the collective modes vector) along the CM is performed. Scaling relations involving the effective diffusion coefficients as well as the characteristic kinetic parameters must be introduced in order to get the adequate smeared Fokker-Planck (FP) equation for the reduced probability $\tilde{P}=\tilde{P}(\mathbf{X}_s,t)$ ($\mathbf{X}_s$ is the subordinating modes vector). That means, to leading order in (W/V) (W is the virtual subvolume, V is the thermodynamic volume), we must obtain a CM-reduced equation which allows for a continuous flow of probability about the CM. In other words, the diffusion pressure must be given by the correlation of fluctuations in the CM slow modes exclusively.[5] This procedure leads to a decomposition of the system in subsystems of subvolume W where

$$(W/V)=L^{-1} , \tag{1}$$

with L^{-1} being the scaling parameter for the correlation of fluctuations in the subordinating modes.

The aim of this work is to show how critical fluctuations propagate and amplify along the CM so that organized states are induced by one subsystem into another. Thus we need to obtain explicitly the time evolution of the probability distribution in a suitable coarse-grained phase space associated to every subsystem.[3,4] The ensemble of subsystems is not an artifact in the Gibbs sense but it is obtained from the fluctuation-correlation scaling which holds when a CM is sustained. On the other hand, the coarse graining is determined by the space of realizations of the random source in the order-parameter equation (cf. Ref. 4). Thus the subsystems associated to macrostates lying in the CM are information carriers or organized subsystems with finite lifetime except those lying in the attractor (in the case of a hard-mode instability, a limit cycle). The subsystems whose macrostate lies in the attractor are information carriers with infinite lifetime. The lifetime of subsystems in a CM and/or attractor is determined by the diffusion pressure about the CM.

We shall define an adequate multiplicity for macrostates not based on equally accessible microstates but based on cells of microstates, so that the coarse graining of phase space thus defined is compatible with the ensemble of realizations of the random source (denoted $\mathbf{f}$). This procedure is carried out by introducing an equivalence relation "$\sim$," where the cells are the equivalence classes. The relation is defined as follows: Fixing arbitrarily a microstate A, all the microstates with the same macrostate as A and connected to A by a phase trajectory with $\Delta\mathbf{f}=0$ are equivalent to A. We denote one such microstate as B. Thus we have

$$A\sim B\leftrightarrow B\in c(A) , \tag{2}$$

where $c(A)$ is the cell containing A. The variation in the source term $\mathbf{f}$ is associated to the displacement along the phase trajectory. Thus the coarse-grained phase space is the space of microstates modulo $\sim$, or the quotient space

$$\bar{\Sigma}=\Sigma/\sim , \tag{3}$$

where Σ denotes the phase space. We need to describe the time evolution of a probability distribution p defined on $\bar{\Sigma}$ determined by the distribution P in the space of collective macroscopic modes. The distribution p is made up of the following thermal averages (the angular brackets represent this thermal average which is in fact the average over the ensemble of subsystems):

$$p=\{p_A\}_{\text{all } c(A)\in\bar{\Sigma}},\quad p_A=\langle\chi_A\rangle \tag{4}$$

where χ_A is the characteristic function for $c(A)$,

$$\chi_A=\begin{cases}1 & \text{if the subsystem is in a microstate}\\ & \text{contained in } c(A)\\ 0 & \text{otherwise .}\end{cases} \tag{5}$$

Thus p_A gives the probability that a subsystem is in cell $c(A)$ at a given time. Let Λ contained in $\bar{\Sigma}$ denote the collection of cells whose macrostates belong to the CM, excluding the attractor emerging beyond a hard-mode instability. A measure of the degree of organization is then given by the fraction of organized subsystems,

$$x=\sum_{c(B)\in\Lambda}\langle\chi_B\rangle . \tag{6}$$

Thus the CM acts as a transient source of free energy since each information carrier has a finite lifetime given by the reciprocal of the effective diffusion coefficient

$$D=S^{-1}\langle\|\mathbf{f}\|^2\rangle^{1/2} , \tag{7}$$

S being the dimension of the CM or the number of order parameters.

The parameter L introduced in Eq. (1) is characteristic of the unfolding and will be used to display the scaling relations involving the following characteristic parameters:

(1) The average Gaussian width of probability density about the CM, denoted $\bar{w}$.

(2) The unfolding or bifurcation parameter, denoted $b=q-q_c$.

(3) The scaling factor for the covariance matrix elements C_{ij}, for the internal fluctuations.

We shall assume, without loss of generality, that the system is already in Poincaré normal form.[3,5] That is, the fast-relaxing subordinated modes $\mathbf{X}_f$ and the enslaving modes $\mathbf{X}_s$ have been separated. In this $(\mathbf{X}_s,\mathbf{X}_f)$ representation, the explicit form of the covariance $\underline{C}$ is given by

$$C_{ij}(t,t')=\langle f_i(t)f_j(t')\rangle=O(L^{-1}) , \tag{8}$$

where the terms f_j are the random sources for slow and fast cooperative modes. We can now outline the CM reduction. Details of this procedure can be found elsewhere.

In general, for S subordinating degrees of freedom and F subordinated variables, with $S+F=N$, we have the following general equation:

$$\int_{\text{c.m.}} \partial_t P(\mathbf{X}_s,\mathbf{X}_f,t)d\mathbf{X}_f = \int_{\text{c.m.}} \left[-\sum_{i=1}^{S} \partial_{X_{s,i}}\{[\dot{X}_{s,i}-(f)_{s,i}]P\} - \sum_{j=1}^{F} \partial_{X_{f,j}}\{[\dot{X}_{f,j}-(f)_{f,j}]P\} \right.$$
$$\left. + \sum_{i,i'=1}^{s} \tfrac{1}{2}\tilde{C}_{ii'}\partial^2_{X_{s,i}X_{s,i'}}P + 2\sum_{i=1}^{S}\sum_{j=1}^{F} \tfrac{1}{2}\tilde{C}_{ij}\partial_{X_{s,i}X_{f,j}}P + \sum_{j,j'=1}^{S} \tfrac{1}{2}\tilde{C}_{jj'}\partial^2_{X_{f,j}X_{f,j'}}P \right] d\mathbf{X}_f \ . \tag{9}$$

In order to integrate this equation, we shall make use of the factorization of the probability functional which gives the statistical subordination of fast variables,

$$P(\mathbf{X}_s,\mathbf{X}_f,t)=\tilde{P}(\mathbf{X}_s,t)Q(\mathbf{X}_f \mid \mathbf{X}_s) \ , \tag{10}$$

$$Q(\mathbf{X}_f \mid \mathbf{X}_s)=\prod_{j=1}^{F} (g_j/\pi)^{1/2}\exp\{-g_j[\mathbf{X}_{f,j}-\tilde{F}_j(\mathbf{X}_s)]^2\} \ . \tag{11}$$

The center manifold hypersurface in macrostate space need not be calculated here; for details, the reader may consult Ref. 5. The CM is given by the equations

$$X_{f,j}=\tilde{F}_j(\mathbf{X}_s) \ . \tag{12}$$

The Gaussian widths about the CM will be derived explicitly together with the scaling relations needed to obtain the smeared FP equation. In general they are given by the following relations:

$$g_j=\sum_{k=0}^{\infty} g_{jk}\left[\prod_{i_1+i_2+\cdots+i_s=k} X_{s,1}^{i_1}X_{s,2}^{i_2}\cdots X_{s,s}^{i_s}\right] \ ,$$
$$w_j=(2g_j)^{-1/2} \tag{13}$$

From now on, the discussion will be centered upon the case of a limit-cycle organization emerging beyond a hard-mode instability with the limit cycle contained in a two-dimensional CM.

We shall now adopt the following scaling relations in order to explicitly display the relative size of the terms resulting from the integration process,

$$C=L^{-1}, \quad X_{s,i}=O(L^{-1/4}) \ ,$$
$$\bar{w}=O(L^{-1/2}), \quad b=O(L^{-1/2}) \tag{14}$$

where $\bar{w}$ is the width of the probability density averaged over the fast degrees of freedom.

For our case of interest, we have

$$\dot{X}_{s,1}-(f)_1=-wX_{s,2}+a_1X_{s,1}^2+b_1X_{s,1}X_{s,2}$$
$$+c_1X_{s,2}^2+O(L^{-3/4}) \ , \tag{15}$$

$$\dot{X}_{s,2}-(f)_2=wX_{s,1}+a_2X_{s,1}^2+b_2X_{s,1}X_{s,2}$$
$$+c_2X_{s,2}^2+O(L^{-3/4}) \ , \tag{16}$$

$$\dot{X}_f-(f)_f=-\lambda X_f+a_{f1}X_{s,1}^3+a_{f2}X_{s,2}^3+O(L^{-1}) \ . \tag{17}$$

Following the canonical procedure for a Hopf instablity, we introduce the cylindrical coordinates

$$X_f=X_f, \quad X_{s,1}=r\cos\theta, \quad X_{s,2}=r\sin\theta \ . \tag{18}$$

Thus we get the following smeared FP equation:

$$\partial_t\tilde{P}=-r^{-1}\partial_r\{[(q-q_c)L^{-1/2}r^2+a_1r^4]\tilde{P}\}-\partial_\theta[(w+b_1r^2)\tilde{P}]$$
$$+[(\tfrac{1}{2}\tilde{C}_{s,1;s,1})^{1/2}\cos\theta+(\tfrac{1}{2}\tilde{C}_{s,2;s,2})^{1/2}\sin\theta]^2\partial^2_{rr}\tilde{P}$$
$$+[-\tfrac{1}{2}\tilde{C}_{s,1;s,1}\cos\theta\sin\theta+(\tilde{C}_{s,1;s,1}\tilde{C}_{s,2;s,2})^{1/2}\cos^2\theta+\tfrac{1}{2}\tilde{C}_{s,2;s,2}\sin\theta\cos\theta]\partial_r(r^{-1}\partial_\theta\tilde{P})$$
$$+r^{-1}[\tfrac{1}{2}\tilde{C}_{s,1;s,1}\sin^2\theta-(\tilde{C}_{s,1;s,1}\tilde{C}_{s,2;s,2})^{1/2}\cos\theta\sin\theta+\tfrac{1}{2}\tilde{C}_{s,2;s,2}\cos^2\theta]\partial_r\tilde{P}$$
$$+r^{-2}[-\tilde{C}_{s,1;s,1}\sin\theta\cos\theta+2(\tilde{C}_{s,1;s,1}\tilde{C}_{s,2;s,2})^{1/2}\sin^2\theta-\tilde{C}_{s,2;s,2}\cos\theta\sin\theta]\tfrac{1}{2}\partial_\theta\tilde{P} \ , \tag{19}$$

where the tilde on top of the correlation matrix elements denotes restriction to the CM. This equation has been obtained making use of the first-order approximation

$$\bar{w}=N^{-1}\sum_{j=1}^{N}(2\tilde{C}_{jj}/\lambda_j)^{1/2} \ , \tag{20}$$

where the λ_j are the damping constants for the $X_{f,j}$ and the subindex f has been dropped from the correlation elements.

Following a standard procedure, we shall get rid of the angular dependence by factorizing $\tilde{P}$ as follows:

$$\tilde{P}(r,\theta,t)=\tilde{\tilde{P}}(r,t)y(\theta \mid r) \ . \tag{21}$$

This gives, to order $L^{-1/2}$, the equation of continuity for

$\tilde{\tilde{P}}$ which ensures the continuous flow of probability about the CM and also verifies the validity of relations (8) and (14),

$$\partial_t \tilde{\tilde{P}}(r,t) = -\frac{1}{r}\partial_r\{[(q-q_c)L^{-1/2}r^2 + ar^4]\tilde{\tilde{P}}\} + \tfrac{1}{4}(\tilde{C}_{s,1;s,1} + \tilde{C}_{s;2;s,2})\partial_{rr}^2 \tilde{\tilde{P}} \ . \quad (22)$$

The decomposition of the system in interacting subsystems of volume W follows from Eqs. (1), (8), and (14).

In order to describe the time behavior of p, we need to derive the time evolution of the set of finite-lifetime information carriers, that is, we restrict the density to

$$p_{\mathrm{CM}} = \{p_B\}_{c(B)\in\Lambda} \ . \quad (23)$$

The time-evolution of p_{CM} depends on the cell correlations

$$S_{AB}(t) = \langle \langle \chi_A(\alpha)\chi_B(\beta)\rangle_\alpha \rangle_\beta \ , \quad (24)$$

where α and β label subsystems in a generic sense. Thus, for an arbitrary $c(B)\in\Lambda$, we have

$$\langle \dot{\chi}_B \rangle = \left[\sum_{c(A)\in\Lambda} \left[\frac{\partial}{\partial t} S_{AB} \right] (1-x)\langle \chi_A \rangle \right] - N^{-1}\langle \|\mathbf{f}\|^2 \rangle^{1/2} \langle \chi_B \rangle \ . \quad (25)$$

The terms proportional to $(1-x)$ give the probability per unit time that a subsystem in a cell $c(A)\in\Lambda$ induces a subsystem outside the CM to become organized by evolving to $c(B)\in\Lambda$. This is so since the probability that a subsystem is in a disorganized cell is $(1-x)$. The remaining term in Eq. (25) corresponds to the destruction of the information carrier. The variable $x(t)$ represents the level of self-organization within the CM. Its behavior depends implicitly on the kinetic parameters since the parameters L, D, and S_{AB} are determined by the CM reduction. The induction period is given by the length of time which must elapse in order for the system to evolve along the CM until the microstates realizing the attractor are reached by all the subsystems. In other words, the induction period is the time it takes the fluctuations to propagate along the CM. Thus it is determined by the time evolution of x. The Perron number (the largest real positive eigenvalue) λ_{Pe} for the matrix $[(\partial/\partial t)S_{AB}]$ coincides with the effective diffusion coefficient D since the final stationary state x_{ss} in the evolution of x is

$$x_{\mathrm{ss}} = (1 - D/\lambda_{\mathrm{Pe}}) = 0 \ . \quad (26)$$

This relation must hold in order for the attractor to be reached by all subsystems after the critical fluctuations have propagated and amplified through the CM. The induction period T_{ind} is given by

$$x(T_{\mathrm{ind}}) = x_{\mathrm{ss}} = 0 \ . \quad (27)$$

In a simplified model we can impose the restriction of equal induction probabilities per unit time between any pair of microstate cells $c(A)$ and $c(B)$. Since L and D can be obtained from a stochastic CM treatment for a particular unfolding, the probability per unit time can be obtained from the fact that the Perron number is determined by Eq. (26).

III. DISCUSSION

The model given by Eqs. (23)–(27) is isomorphic to Eigen's selection model for biopolymer information carriers.[9] This isomorphism can be made explicit if we adopt the following representation:

$$y_A = p_A/x \ . \quad (28)$$

Then Eq. (25) reads

$$\dot{y}_A = \sum_{c(B)\in\Lambda} M_{AB}y_B - y_A \sum\sum_{c(C),c(B)\in\Lambda} M_{CB}y_B \ , \quad (29)$$

where

$$M_{AB} = (1-x)(\partial/\partial t)S_{AB} - N^{-1}\langle \|\mathbf{f}\|^2 \rangle^{1/2}\delta_{\chi_A\chi_B} \ . \quad (30)$$

In Eigen's selection model, the information carriers are the biopolymers which in our model have been replaced by subsystems lying in the CM. The macromolecular information carriers are subject to mutations with rates $w_{ij}y_i$, for a mutation $i\to j$ (i,j two biopolymers), and autocatalytic self-replication with rate $A_iQ_iy_i$ (where A_i is the rate constant, Q_i the quality factor, and mass-action kinetics is assumed).

If we now impose the constraint of constant overall population ($\sum_i y_i = \mathrm{const}$), thus adjusting the dilution fluxes to account for the metabolic turnover, we have an explicit isomorphism between Eigen's model and the evolution of organized subsystems given by the map: organized subsystem $\to$ macromolecular information carrier (linear biopolymer); $A_i \to i$; $(\partial/\partial t)S_{A_iA_j}(1-x) \to w_{ij}$ (if $i\neq j$), A_jQ_j (otherwise); $D\to D$ (death rate for an information carrier).

The system defined by Eqs. (23)–(27) provides an efficient algorithm for computing lag times required to reach a dissipative structure emerging beyond a hard-mode instability. This is a central problem which to the best of the author's knowledge has received little attention so far. The rates of decay of other metastable states are well determined theoretically as well as experimentally. Instructive examples are the condensation of supersaturated vapor, the nucleation phenomena leading to phase transitions in alloys and magnets, and the amplification of disturbances in superfluids; in this case, the disturbance starts as a vortex ring of critical size.

In order to appreciate the advantages of the representation introduced in this work, let us remark that if we were to proceed following the classical theory of decay of metastable states for the region of propagation of fluctuations along the CM, we would need to consider the time evolution of the distribution $\rho = \rho(\{\eta\})$, where

$$\{\eta\} = \{\eta_1, \eta_2, \ldots, \eta_L\} \quad (31)$$

represents a particular configuration. The order parameter evaluated at subsystem α, η_α ($\alpha = 1,2,\ldots,L$) is defined as

$$\eta_\alpha=\eta(\alpha)=\sum_{c(B)\in\Lambda}\chi_B(\alpha)\ . \tag{32}$$

Thus we get the general master equation for ρ as

$$\left[\frac{\partial\rho\{\eta\}}{\partial t}\right]=\int\delta\eta'[P(\{\eta\},\{\eta'\})\rho\{\eta'\} -P(\{\eta'\},\{\eta\})\rho\{\eta\}]\ , \tag{33}$$

where the coupling to the statistical reservoir is explicitly given by the transition rates between any two given configurations. These rates are denoted P and they have the form

$$P(\{\eta\},\{\eta'\})=\sum_i\prod_{j\neq i}\delta(\eta_j-\eta_j')R_i(\{\eta\},\{\eta'\})\ , \tag{34}$$

where the transition probabilities R_i can be written as

$$R_i=\int ds\int ds'\left[\frac{e^{-\varepsilon(s')/kT}}{Z_R}\right]T_i(s,\eta_i;s',\eta_i') \times\delta[\varepsilon(s)+E\{\eta\}-\varepsilon(s')-E\{\eta'\}]\ , \tag{35}$$

where s and s' denote states of the bath, $\varepsilon(s)$ and $\varepsilon(s')$ the energies for these states, Z_R the partition function for the statistical bath, and $E\{\eta\}$ and $E\{\eta'\}$, the energies of the two configurations, respectively. The reader can readily see that the conservation of energy is satisfied by Eq. (35).

The main difficulty in evaluating the T_i's lies in the fact that the system is *not* evolving to equilibrium. Therefore the T_i's are dependent on the full configurations $\{\eta\}$ and $\{\eta'\}$, since organized subsystems induce phase trajectories into the CM thus organizing disorganized subsystems spatially coupled to them. For this reason, the classical representation, widely used in nucleation phenomena, is to be replaced by the more convenient one as given by Eqs. (23)–(27).

IV. CONCLUSION

We have demonstrated explicitly the role of correlations of spatially coupled subsystems at the onset of a center manifold in open reactors which operate far from equilibrium. Thus such dissipative processes can be viewed in the same context as spinodal decompositions, where the correlation of cells leads to the conservation of the order parameter,[8] or in the amplification of nonequilibrium intrinsic fluctuations in nucleation phenomena. The organizing dynamics for subsystems lying in the center manifold was shown to be rigorously equivalent to the selection process in the self-reproduction of macromolecular information carriers as given in Eigen's model.[9]

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